

Critical magnetization behaviors of the triangular and Kagome lattice quantum antiferromagnets

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We investigate the $S = 1/2$ quantum spin antiferromagnets on the triangular and Kagome lattices in magnetic field, using the numerical exact diagonalization. Particularly we focus on an anomalous magnetization behavior of each system at $1/3$ of the saturation magnetization. The critical exponent analyses suggest that it is a conventional magnetization plateau on the triangular lattice, while an unconventional phenomenon, called the magnetization ramp, on the Kagome lattice.

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The $S = 1/2$ triangular and Kagome¹ lattice antiferromagnets have attracted a lot of interests as typical frustrated systems. Most theoretical studies indicated that the former system has the three sublattice long-range order²⁻⁴, while the latter is disordered in the ground state⁵⁻¹⁶. Experimental studies to observe a novel spin liquid phase have been accelerated since discoveries of several realistic materials; the organic compound κ -(BEDT-TTF)₂Cu₂(CN)₃ for the triangular lattice¹⁷, the herbertsmithite^{18,19}, the volborthite^{20,21} and the vesignieite²² for the Kagome lattice. Since the quantum Monte Carlo simulation and the DMRG calculation are useless for these systems, the numerical exact diagonalization is one of the best numerical method for them. The numerical diagonalization studies suggested that both systems have the $1/3$ magnetization plateau²³⁻²⁷, although the classical spin systems have no plateau on both lattices in the ground state^{28,29}. (The thermal or quantum fluctuations induce a plateau in the semiclassical case, because $1/3$ is just a critical point between two different spin structures.) In our recent numerical diagonalization study on the $S = 1/2$ Kagome lattice antiferromagnet up to $N = 36$, the calculated field derivatives reveal an anomalous behavior at $1/3$ of the saturation magnetization³⁰. Namely, the field derivative is diversing at the low-field side of the critical field H_c , while almost zero at the high-field side. This critical behavior is quite different from conventional magnetization plateaux in two-dimensional systems where the field derivative is finite at both sides of H_c . To distinguish such an anomalous property at the $1/3$ magnetization of the Kagome lattice from conventional plateaux, we called it a “magnetization ramp”. However, its mechanism is still an open problem. In this paper, to clarify such an unconventional behavior around the $1/3$ magnetization of the $S = 1/2$ Kagome lattice antiferromagnet, comparing with the triangular one, we applied the numerical diagonalization for both systems up to $N = 39$ which is the largest cluster at present. In addition we estimated the

critical exponent δ by the finite-size scaling proposed by the previous work³², to investigate the quantum critical behavior more quantitatively.

Now we examine the magnetization process of the $S = 1/2$ triangular and kagome lattice antiferromagnets. The Hamiltonian is given by

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_Z, \quad (1)$$

$$\mathcal{H}_0 = \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \quad \mathcal{H}_Z = -H \sum_j^N S_j^z, \quad (2)$$

where $\langle i, j \rangle$ means all the nearest neighbor pairs on each lattice. Throughout we use the unit such that $g\mu_B = 1$. For N -site systems, the lowest energy of \mathcal{H}_0 in the subspace where $\sum_j S_j^z = M$ (the macroscopic magnetization is $m = M/M_s$, where M_s denotes the saturation of the magnetization, namely $M_s = NS$ for the spin- S system.) is denoted as $E(N, M)$. We restrict us to the rhombic cluster under the periodic boundary condition to keep the 120° rotational symmetry for a systematic finite-size scaling. Using the numerical exact diagonalization, we have calculated all the values of $E(N, M)$ available for the rhombic clusters with $N = 9, 12, 21, 27, 36$ and 39 , to obtain the ground state magnetization curves. (The largest dimension of the $N = 39$ system is 68,923,264,410. To treat such huge matrices in computers, we have carried out parallel calculations using the MPI-parallelized code which is originally developed in the previous work³¹.) The magnetization curves of the (a) triangular and (b) Kagome lattice antiferromagnets are shown in Fig. 1 for $N = 27, 36$ and 39 . They indicate plateau-like behaviors at $m = 1/3$ of both systems, but the Kagome lattice exhibits an anomalous feature; the step length increases with decreasing H towards $m = 1/3$, different from conventional magnetization plateau like the triangular lattice. In order to clarify a difference between the triangular and Kagome lattice, we calculated the field

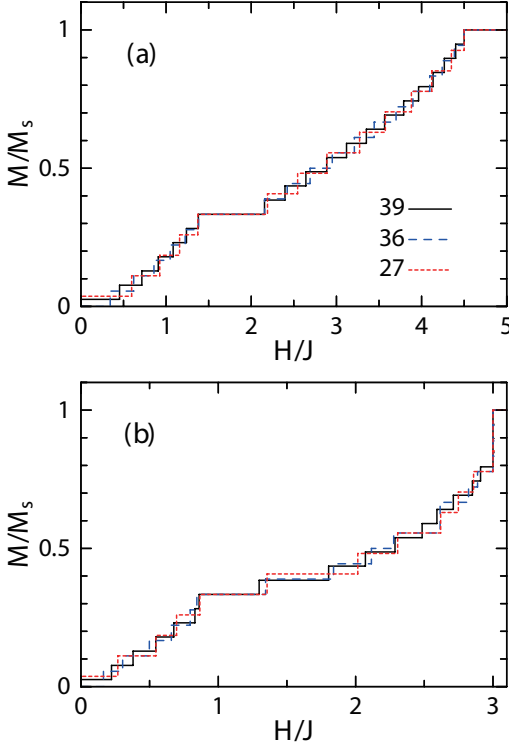


FIG. 1: Magnetization curves of the (a) triangular and (b) Kagome lattice antiferromagnets for $N = 27, 36, 39$.

derivative χ defined the form

$$\chi^{-1} = \frac{E(N, M+1) - 2E(N, M) + E(N, M-1)}{1/M_s}. \quad (3)$$

The derivative χ of the (a) triangular and (b) Kagome lattice systems are shown in Fig. 2 for $N = 27, 36$ and 39 . The derivative χ of the triangular system is finite at both edges of the $1/3$ plateau-like behavior, as well as conventional two-dimensional systems. In contrast, the Kagome system exhibits a quite different feature between the lower and higher field sides of $m = 1/3$; χ is diverging at the lower side like a plateau in one-dimensional systems, while is very small (possibly zero) at the higher one. The present calculation for $N = 39$ more strongly supports a ramp-like behavior predicted by our previous work.

The critical exponent δ defined by the form

$$|m - m_c| \sim |H - H_c|^{1/\delta}, \quad (4)$$

is an important index to specify the universality class of the field induced quantum phase transition. The previous theoretical works indicated $\delta = 2$ for some typical one-dimensional gapped systems^{33,34}, while $\delta = 1$ for two-dimensional systems³⁵. In order to investigate the quantum critical behavior at $m = 1/3$ of the triangular and Kagome lattice antiferromagnets more quantitatively, we estimate δ by the finite-size scaling developed by the previous work³². Although it was proposed for

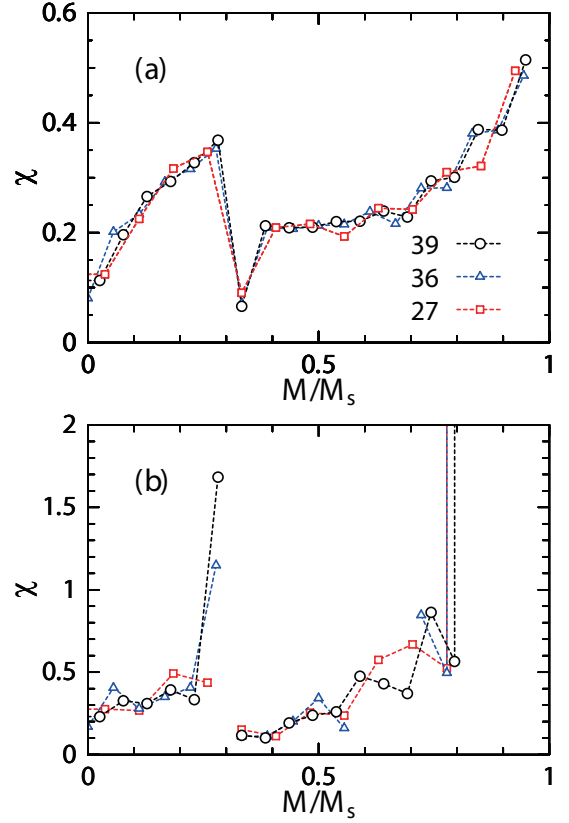


FIG. 2: Field derivatives of the (a) triangular and (b) Kagome lattice antiferromagnets for $N = 27, 36, 39$.

one-dimensional systems, it can be easily generalized for two dimensions. We assume the asymptotic form of the size dependence of the energy as

$$\frac{1}{N}E(N, M) \sim \epsilon(m) + C(m)\frac{1}{N^\theta} \quad (N \rightarrow \infty), \quad (5)$$

where $\epsilon(m)$ is the bulk energy and the second term describes the leading size correction. We also assume that $C(m)$ is an analytic function of m . The lowest and highest magnetic field corresponding to $m = 1/3$ in the thermodynamic limit are defined as H_{c-} and H_{c+} , respectively, as the form

$$E(N, \frac{N}{3} \pm 1) - E(N, \frac{N}{3}) \rightarrow \pm H_{c\pm} \quad (N \rightarrow \infty). \quad (6)$$

In order to consider the critical magnetization behaviors for $m < 1/3$ and $m > 1/3$ independently, we define the critical exponents δ_{c-} and δ_{c+} by the forms

$$|m - \frac{1}{3}| \sim |H - H_{c\pm}|^{1/\delta_{\pm}}. \quad (7)$$

If we define the quantities $f_+(N)$ and $f_-(N)$ by the forms

$$\begin{aligned} f_{\pm}(N) \\ \equiv \pm[E(N, \frac{N}{3} \pm 2) + E(N, \frac{N}{3}) - 2E(N, \frac{N}{3} \pm 1)], \end{aligned} \quad (8)$$

the asymptotic forms of them are expected to be

$$f_{\pm}(N) \sim \frac{1}{N^{\delta_{\pm}}} + O\left(\frac{1}{N^{\theta+1}}\right) \quad (N \rightarrow \infty), \quad (9)$$

as far as we assume the form (6). Thus the exponents δ_- and δ_+ can be estimated from the slope of the $\ln f_{\pm}$ - $\ln N$ plot, respectively, under the condition $\theta > \delta_{\pm} - 1$. In order to avoid an oscillation of the finite-size correction due to the cluster shape dependence, we just use the rhombic clusters under the periodic boundary condition with $N = 9, 12, 21, 27, 36$, and 39 . The plots of $\ln f_{\pm}$ versus $\ln N$ for the (a) triangular and (b) Kagome lattice antiferromagnets are shown in Fig. 3. Figure 3 (a) suggests that the calculated points are well fitted to a line for each of f_- and f_+ in the case of the triangular lattice. Thus applying the standard least square fitting to lines (Dashed and long-dashed lines are used to obtain δ_+ and δ_- , respectively.) for all the available system sizes; $N = 9, 12, 21, 27, 36, 39$ ($N = 9$ cannot be used for δ_-), δ_- and δ_+ are estimated as follows:

$$\delta_- = 1.00 \pm 0.17, \quad \delta_+ = 0.89 \pm 0.15,$$

for the triangular lattice. The errors are estimated from the deviation of points from the fitted lines. It would be reasonable to conclude $\delta_- = \delta_+ = 1$ at $m = 1/3$ of the triangular lattice antiferromagnet, as expected for conventional magnetization plateau in two dimensions. On the other hand, Fig. 3 (b) indicates quite different feature of the Kagome lattice antiferromagnet. The same least square fitting yields the following estimates:

$$\delta_- = 1.92 \pm 0.99, \quad \delta_+ = 0.56 \pm 0.15,$$

for the Kagome lattice antiferromagnet. Exponent δ_- has a large error because the line fitting is not good. It does not converge with respect to the system size well, but seems to still increasing with N . The same line fitting to the points for $N = 27, 36$ and 39 yields the estimation $\delta_- = 4.59 \pm 0.25$. Thus we can just conclude $\delta_- \geq 2$ at most. It means that the diversing behavior of the field derivative at H_{c-} is stronger than one-dimensional systems. It is led to two possibilities. One is a jump (a first-order transition) in the magnetization curve. A magnetization jump which also appears near the saturation was proved³⁶. The other is an anomalous continuous transition. A similar phenomenon was reported in the metal-insulator transition of the Hubbard chain with next-nearest neighbor hopping³⁷. In comparison with δ_- , δ_+ is more conclusive, because the fitting error is much smaller. According to the above result of the line fitting, we conclude $\delta_+ = 0.6 \pm 0.2$. Thus the field derivative χ should be zero at the higher field side of H_{c+} , because δ_+ is smaller than unity. It also justifies a property of the magnetization ramp.

Finally, we consider whether a flat part of the magnetization curve at $m = 1/3$ exists or not for the triangular and Kagome lattice antiferromagnets. Namely, we examine whether each system has no plateau ($H_{c-} = H_{c+}$) or

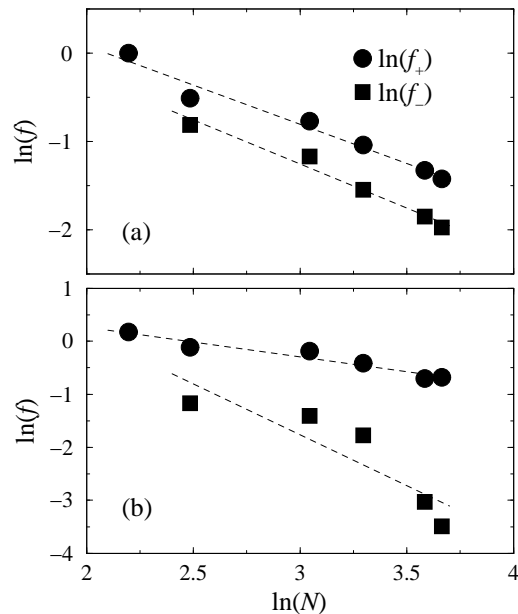


FIG. 3: $\ln(f)$ is plotted versus $\ln(N)$ for (a) the triangular and (b) Kagome lattice antiferromagnets, respectively. Solid circles and squares are useful to estimate the critical exponents δ_+ and δ_- , respectively.

a finite plateau ($H_{c-} \neq H_{c+}$) at $m = 1/3$ in the thermodynamic limit. We evaluate the length of the flat part $H_{c+} - H_{c-}$ corresponding to the plateau width of the finite-size clusters with $N = 9, 12, 21, 27, 36$ and 39 for both systems. If the system has a gapless excitation like a spin wave from some ordered states, the low-lying energy spectrum is expected to be proportional to the wave vector k in the long wave length limit. Thus the excitation energy gap of the finite-size systems should have the asymptotic form $\sim 1/N^{1/2}$ in two-dimensional gapless systems. On the other hand, in gapped systems the gap is expected to converge to the thermodynamic limit with exponentially decaying (faster than $1/N^{1/2}$) finite-size correction, as the system size increases. Thus if the extrapolation by fitting the gap versus $1/N^{1/2}$ leads to a finite gap in the thermodynamic limit, it would be a strong evidence to confirm the gapped ground state. The length of a flat part $H_{c+} - H_{c-}$ is plotted versus $1/N^{1/2}$ in Fig. 4, where open triangles and solid circles are for the triangular and Kagome lattice antiferromagnets, respectively. The least square fitting to a line leads to the following results: $H_{c+} - H_{c-} = 0.47 \pm 0.28$ for the triangular lattice and $H_{c+} - H_{c-} = -0.32 \pm 0.35$ for the Kagome lattice. Obviously we can conclude that the triangular lattice antiferromagnet has the $1/3$ magnetization plateau. In contrast, the result for the Kagome lattice suggests that it possibly has a single critical field $H_c = H_{c-} = H_{c+}$. However, it is difficult to exclude a finite magnetization plateau, because of a large error of the extrapolation. Note that any other plateaux are difficult to investigate by the present method, because fewer

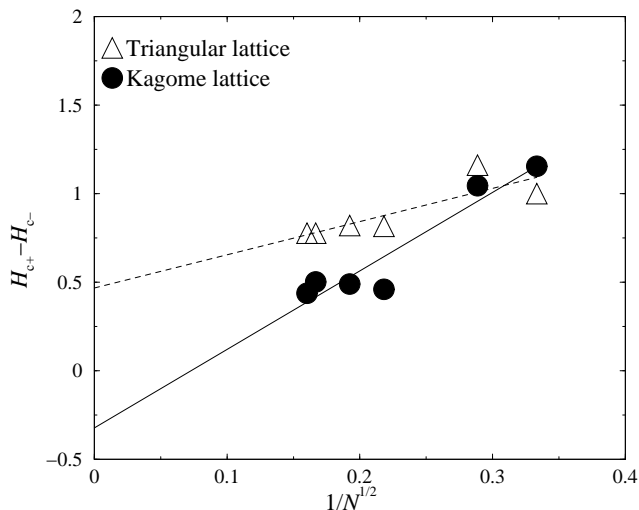


FIG. 4: Plateau width $H_{c+} - H_{c-}$ is plotted versus $1/N^{1/2}$. Open triangles and solid squares are for the triangular and Kagome lattice antiferromagnets, respectively. Fitted lines are used for the extrapolation to the thermodynamic limit.

system sizes can be available for $m \neq 1/3$.

In the recent magnetization measurement²⁰ on a candidate of the Kagome lattice antiferromagnet Volborthite several step-like behaviors were observed, but it has not reached $m = 1/3$ yet. The same measurement is still going on to observe an anomaly at $m = 1/3$, which is expected to be about 60T. It would be interesting to detect some unconventional features.

In summary, we have investigated critical magnetiza-

tion behaviors at $m = 1/3$ for the $S = 1/2$ triangular and Kagome lattice quantum antiferromagnets, using the numerical exact diagonalization of rhombic clusters up to $N = 39$. The triangular lattice is revealed to have the critical exponents $\delta_- = \delta_+ = 1$ and a finite plateau, which are consistent with a conventional magnetization plateau in two-dimensional systems. On the other hand, the Kagome lattice is revealed to exhibit unconventional critical properties; $\delta_- < 1 < \delta_+$, namely the field derivative χ is diverging at the lower field side, while zero at the higher one of a possibly single critical field $H_c = H_{c-} = H_{c+}$. The conclusion supports the magnetization ramp behavior at $m = 1/3$ of the Kagome lattice antiferromagnet.

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